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Key Points:

- A summertime Rocky Mountain STE event has been illustrated with high-resolution measurements
- A 10–30 ppbv increase in tropospheric ozone at Fort Collins, CO, is related to stratospheric air
- Stratospheric air is responsible for 18–31% of tropospheric ozone at Fort Collins during 2012–2014

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Characterizing the lifetime and occurrence of stratospheric-tropospheric exchange events in the rocky mountain region using high-resolution ozone measurements

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Abstract The evolution of a Stratospheric-Tropospheric Exchange (STE) event from 4 to 8 August 2014 at Fort Collins, Colorado, is described. The event is characterized with observations from the Goddard Space Flight Center TROPospheric OZone (TROPOZ) Differential Absorption Lidar, the University of Wisconsin High Spectral Resolution Lidar, and multiple ozonesondes during NASA's Deriving Information on Surface Conditions from Column and Vertically Resolved Observations Relevant to Air Quality and the Front Range Air Pollution and Photochemistry Experiment (FRAPPE) campaigns. Based on the extended TROPOZ observations throughout the entire campaign, it was found that STE events have largely contributed to an additional 10–30 ppbv of ozone at Fort Collins. Additional measurements of ozone and relative humidity from the Atmospheric Infrared Sounder are characterize the transport of the intrusion. The Real-time Air Quality Modeling System simulated ozone agrees well with the TROPOZ ozone concentrations and altitude during the STE event. To extend the analysis into other seasons and years, the modeled ozone to potential vorticity ratio is used as a tracer for stratospheric air residing below the tropopause. It is found that at Fort Collins, CO, and depending on season from 2012 to 2014, between 18 and 31% of tropospheric ozone corresponds to stratospheric air. A relationship to determine the lifetime of stratospheric air below the tropopause is derived using the simulated ratio tracer. Results indicate that throughout summer 2014, 43% of stratospheric air resided below the tropopause for less than 12 h. However, nearly 39% persisted below the tropopause for 12–48 h and likely penetrated deeper in the troposphere.

1. Introduction

Intrusions of stratospheric air into the troposphere and tropopause folds are typically prompted by upper level synoptic meteorology such as baroclinic waves and upper level frontogenesis in the jet stream region [*Danielsen*, 1968]. The frontogenetically induced ageostrophic cyclonic circulation forces stratospheric air down into the troposphere, resulting in a folding of the tropopause [*Beekmann et al.*, 1997]. For the midlatitude Rocky Mountain region, stratosphere-troposphere exchanges (STE) and tropopause folds are typically induced by the formation of an upper level cutoff low-pressure system and its associated trough in the presence of a polar front or subtropical jet stream [*Langford*, 1999].

During STE and tropopause fold events, cold, dry, and ozone-rich air are mixed down, sometimes irreversibly, from the stratosphere into the upper free troposphere. If the stratospheric intrusion is dynamic enough, it may transport ozone-rich air directly into the boundary layer [*Davies and Schuepbach*, 1994]. Because deeper intrusions often reside in the troposphere below the climatological tropopause for a longer duration [*Stohl et al.*, 2003], it is important to quantify the seasonal occurrence of STE events, as well as their lifetime.

Several measurements have been made in instances directly involved in STE events or tropopause folds in the Northern Hemisphere in winter [*Rao et al.*, 2003], spring [*Browell et al.*, 1987; *Kuang et al.*, 2012], fall

[*Ancellet et al.*, 1994; *Langford et al.*, 1996; *Sullivan et al.*, 2014], and summer [*Thompson et al.*, 2007; *Yorks et al.*, 2009; *Martins et al.*, 2013]. However, these analyses may be further extended with continuous observations from multiple lidar and balloon-borne measurements, particularly with instruments that have the ability to characterize the atmosphere from near the surface to above the tropopause. An interesting example of a summertime STE event occurred during the concurrent 2014 Deriving Information on Surface Conditions from Column and Vertically Resolved Observations Relevant to Air Quality (DISCOVER AQ) and Front Range Air Pollution and Photochemistry Experiment (FRAPPE) campaigns.

During the observed STE event, the transfer of dry, cold, and ozone-rich stratospheric air was associated with the presence of an Aleutian Low pressure system and developing cutoff low near the Pacific coast of California. The filament of stratospheric air was quickly advected over the Pacific Southwest United States by a persistent southwesterly upper level tropospheric jet stream and eventually transported over the Rocky Mountain region. One unique feature of this STE event is that wildfire aerosols from Southern California were jointly advected below the stratospheric high-ozone air mass to the Rocky Mountain region and are used as an additional tracer of the dynamics of the event.

Because of the campaign, high-resolution ozone lidar measurements from the Goddard Space Flight Center TROPospheric OZone Differential Absorption Lidar (GSFC TROPOZ DIAL) and ozonesondes were available to quantify the upper level ozone structure and illustrate the dynamics surrounding the event. Along with these and other detailed supporting aerosol backscatter observations from the University of Wisconsin High Spectral Resolution Lidar (HSRL), the first part of this paper characterizes the intrusion and advection of a stratospheric air mass over the Rocky Mountain region between 4 and 8 August 2014. Specifically, it is evident from the extended ozone lidar measurements during the DISCOVER AQ campaign that an increase in ozone between 10 and 30 ppbv throughout July and August 2014 in the upper free troposphere at Fort Collins, CO, is largely a contribution from stratospheric air.

In further support of the DISCOVER AQ and FRAPPE campaigns, model assimilations from the Real-time Air Quality Modeling System (RAQMS) were initialized daily. From 4 to 8 August 2014, RAQMS simulated an ozone filament which was similar in altitude and concentration to the TROPOZ and ozonesonde observations. To extend these analyses, the latter portion of this paper uses RAQMS model output, specifically the ozone to potential vorticity ratio, to investigate the stratospheric contribution to Fort Collins and the Rocky Mountain region. It was found that stratospheric ozone residing in the free troposphere is responsible from at least 18–31% of all tropospheric ozone, depending on the season throughout 2012–2014. Results focusing on the summer 2014 indicate that 43% of stratospheric air resided below the tropopause for less than 12 h, corresponding to rather shallow events. However, 38% lasted 12–48 h below the tropopause, implying more than a third of the STE events are related to multiday events.

2. Observations, Retrievals, and Model Output

2.1. Lidar Observations During the 6–7 August 2014 STE Study

The mobile ground-based GSFC TROPOZ DIAL has been routinely taking measurements in the Baltimore-Washington DC, region since fall of 2013 as part of the Tropospheric Ozone Lidar Network (TOLNet). Many of the instrument and retrieval specifications, along with the characterization of an STE event in Maryland can be found in *Sullivan et al.* [2014]. The TROPOZ system was installed in a 13 m transportable trailer which was deployed to Fort Collins, CO (40.59°N, −105.14°W, 1569 m above sea level (asl)). Prior to the deployment, TROPOZ was calibrated against another TOLNet lidar, the Langley Mobile Ozone Lidar, and multiple ozonesondes [*Sullivan et al.*, 2015a].

The TROPOZ observations (Figure 1a) indicate a STE event due to the high concentration (100–150 ppbv) of stratospheric ozone subsiding below the tropopause and entering the free troposphere, the anomalously low concentration (20–40 ppbv) of ozone residing above the high-ozone region, and the increased tropopause height [*Danielsen*, 1968; *Holton et al.*, 1995]. The red triangle at 16:46 UTC is an overlay of the ozone profile from an ozonesonde launched from the Fort Collins, CO, site and is mostly within 10–15% of the TROPOZ retrieval, which is consistent with the results in *Sullivan et al.* [2015b]. The ozone tropopause height is mostly centered around 15.5 km throughout the lidar observation period and is denoted by the yellow triangles throughout the observation. These are derived from the *Bethan et al.* [1996] definition of the altitude in which the ozone mixing ratio is greater than 80 ppbv, ozone gradient exceeds 60 ppbv/km in a depth of 200 m, and the ozone above the tropopause exceeds 110 ppbv.

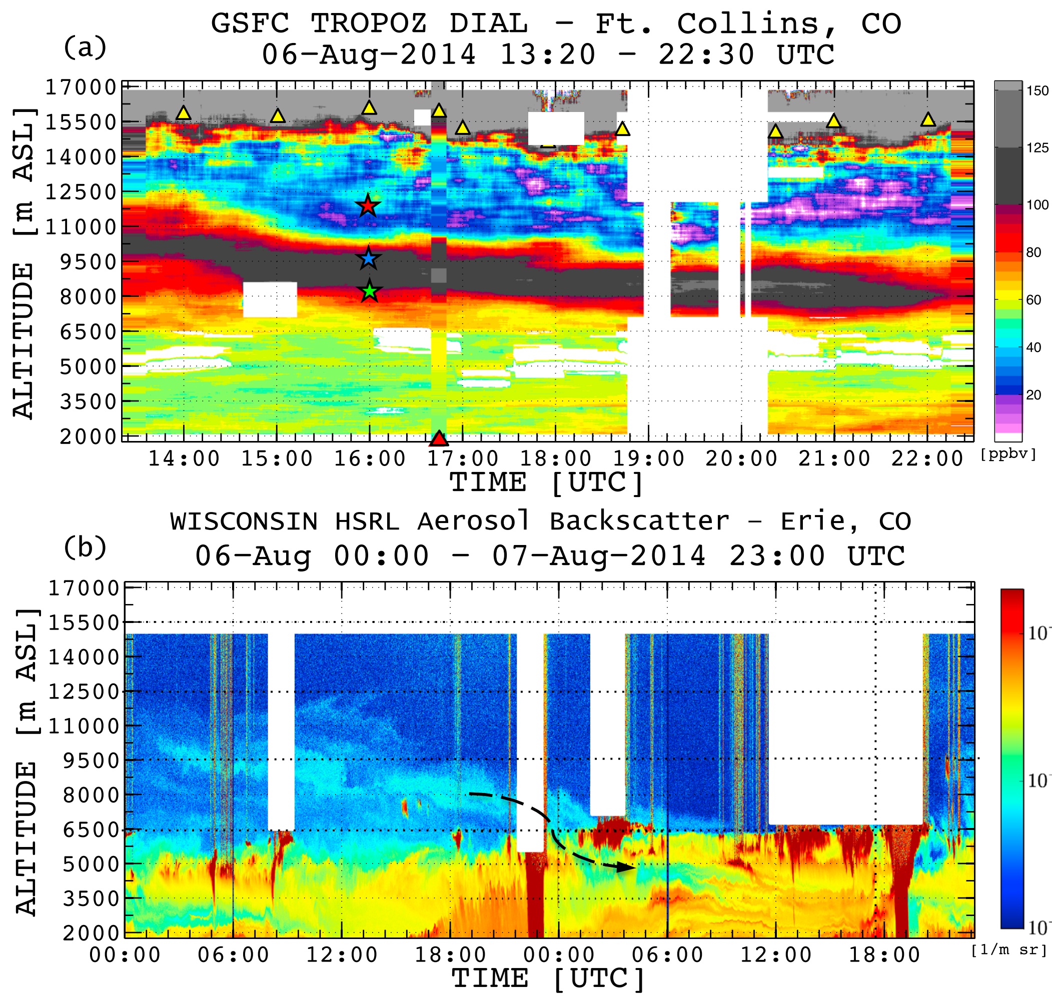

Figure 1. (a) Retrieved TROPOZ ozone concentrations at 30 min resolution from 13:20 to 22:30 UTC on 6 August 2014. The red triangle at 16:46 UTC is the ozonesonde profile, and the yellow triangles indicate the calculated tropopause heights as defined by *Bethan et al.* [1996] at hourly intervals. The stars positioned on the TROPOZ observations indicate the starting time and altitudes of the back trajectory performed in Figure 3. The white regions are periods of the retrieval that were influenced by clouds or regions of insufficient signal. (b) Aerosol backscatter profiles from the University of Wisconsin High Spectral Resolution Lidar (HSRL) from 6 to 7 August 2014 from 0:00 to 23:00 UTC. The black dashed line indicates the downward motion of the aloft aerosol.

The TROPOZ observations indicate that the stratospheric ozone reservoir existed before 13 UTC on 6 August 2014 and suggest transport into the free troposphere. Vertical mixing of the atmosphere above and below the intrusion is minimal, and a region of anomalously low ozone lies just above the stratospheric air mass. These low-ozone concentrations, which were also observed by the ozonesonde, are attributed to a tropical marine air mass via a trajectory assessment (Figure 3), and the blue, red, and green stars in Figure 1a refer to the starting altitudes and locations.

The corresponding HSRL time series (Figure 1b) indicates a perturbation of nearly $10\frac{1}{\mathrm{m\,sr}}$ in the aerosol backscatter centered around 8.5 km. Near 6 August 14 UTC, the observations of the HSRL aerosol perturbation lie underneath the observations of the ozone structure from the TROPOZ and are likely elevated due to the downward motion of the air flow around the jet. For this reason, the HSRL observations are used as a first-order approximation of the dynamics of the STE event where no TROPOZ observations are available. Near 0 UTC on 7 August 2014, the air mass descends to near 6500 m asl and continues in a downward motion toward the planetary boundary layer (PBL). It appears as if this aerosol layer, which has substantially less backscatter than the remaining aerosols within the PBL, continues to descend and impacts the PBL (denoted by black arrow). By 12 UTC, these layers appear to be more well mixed within the PBL. The event likely ends near 18 UTC as precipitation occurs and removes a significant portion of the aerosol from the PBL.

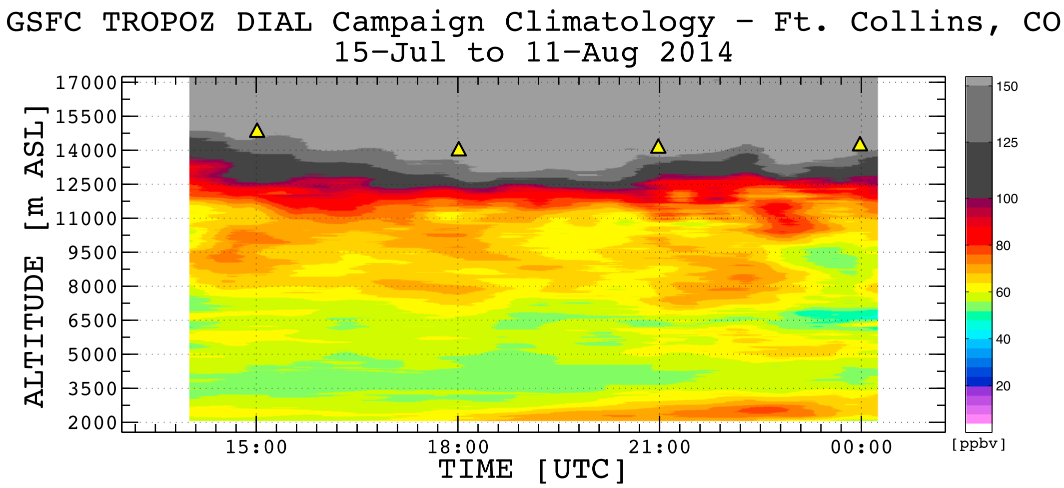

Figure 2. The average TROPOZ time series of over 200 h of daytime and nighttime observations on 18 separate days of retrieved ozone profiles from July to August 2014 during the DISCOVER AQ mission. The yellow triangles indicate the calculated tropopause heights as defined by *Bethan et al.* [1996] at 3 h intervals. For the DISCOVER AQ campaign, TROPOZ observations indicate an increase in upper free tropospheric ozone concentrations, between 10 and 30 ppbv, as a direct contribution from recent stratospheric influx.

2.2. Seasonal Observations of Stratospheric Influence During DISCOVER AQ 2014

Figure 2 illustrates the vertical distribution of ozone and residual stratospheric air that was observed at Fort Collins, CO. It can be interpreted as a "campaign climatology" of the daytime evolution of tropospheric ozone from approximately 2 to 17 km asl at 30 min temporal resolution. Yellow triangles indicate the calculated tropopause heights as defined by *Bethan et al.* [1996] at a 3 h interval. Altitudes that had two or fewer days of data were removed for this figure, ensuring an average of at least 3 days was performed for each range bin. The observations during the high ozone 6–7 August STE case study were removed because peak concentrations were 25–50 ppbv higher than most other days. To ensure the removal of potential biases from aerosols in the final climatology, portions of ozone profiles were removed that corresponded to optically thick wildfire plumes. After the mean profiles were calculated, a five-point temporal and vertical moving average was performed.

Below 3.5 km in Figure 2, photochemically produced ozone has peak concentrations near 60 ppbv at 18:00 UTC (local noon) and steadily grows throughout the day with concentrations centered around 70 ppbv. Near 22:30 (local 4:30 P.M.) there is an increase of roughly 10 ppbv in ozone that presumably comes from increased photochemical production and precursor emissions attributed to mobile sources or local transport from the Denver metropolitan area. In the free troposphere, although the causes and responses to the baseline of background ozone are highly debated [*Reid et al.*, 2008; *Cooper et al.*, 2015], the average concentrations appear to be between 55 and 65 ppbv. Furthermore, during the campaign, RAQMS model output estimates of lighting NO_x production accounted for less than 1 ppbv/d of ozone [*Pierce et al.*, 2007]. The largest increases in ozone concentration occur within the first 2 km below the tropopause, which are likely indicative of shallow stratospheric events. However, there are increases in ozone concentrations that are observed much deeper into the lower free troposphere. Based on this calculated background value throughout the DISCOVER AQ campaign, TROPOZ observations indicate increases in free tropospheric ozone concentrations at Fort Collins, between 10 and 30 ppbv, are largely a contribution from stratospheric influx.

2.3. Back Trajectory

In Figure 3a, the initialized red trajectory altitude (denoted by the star in Figure 1a at 16 UTC) was chosen as a center point of the low-ozone region, and the blue and green altitudes were specifically chosen as boundaries of the high-ozone region. The red trajectory is indicative of an air parcel that was lifted from the tropical marine boundary layer and advected poleward toward the continental United States. The blue trajectory denotes an air parcel that maintained a mostly westerly transport in the troposphere over the Pacific Ocean. The green trajectory is indicative of an air parcel that was cyclonically rotated near the Aleutian Low. The National Centers for Environmental Prediction (NCEP)/National Center for Atmospheric Research (NCAR) Reanalysis

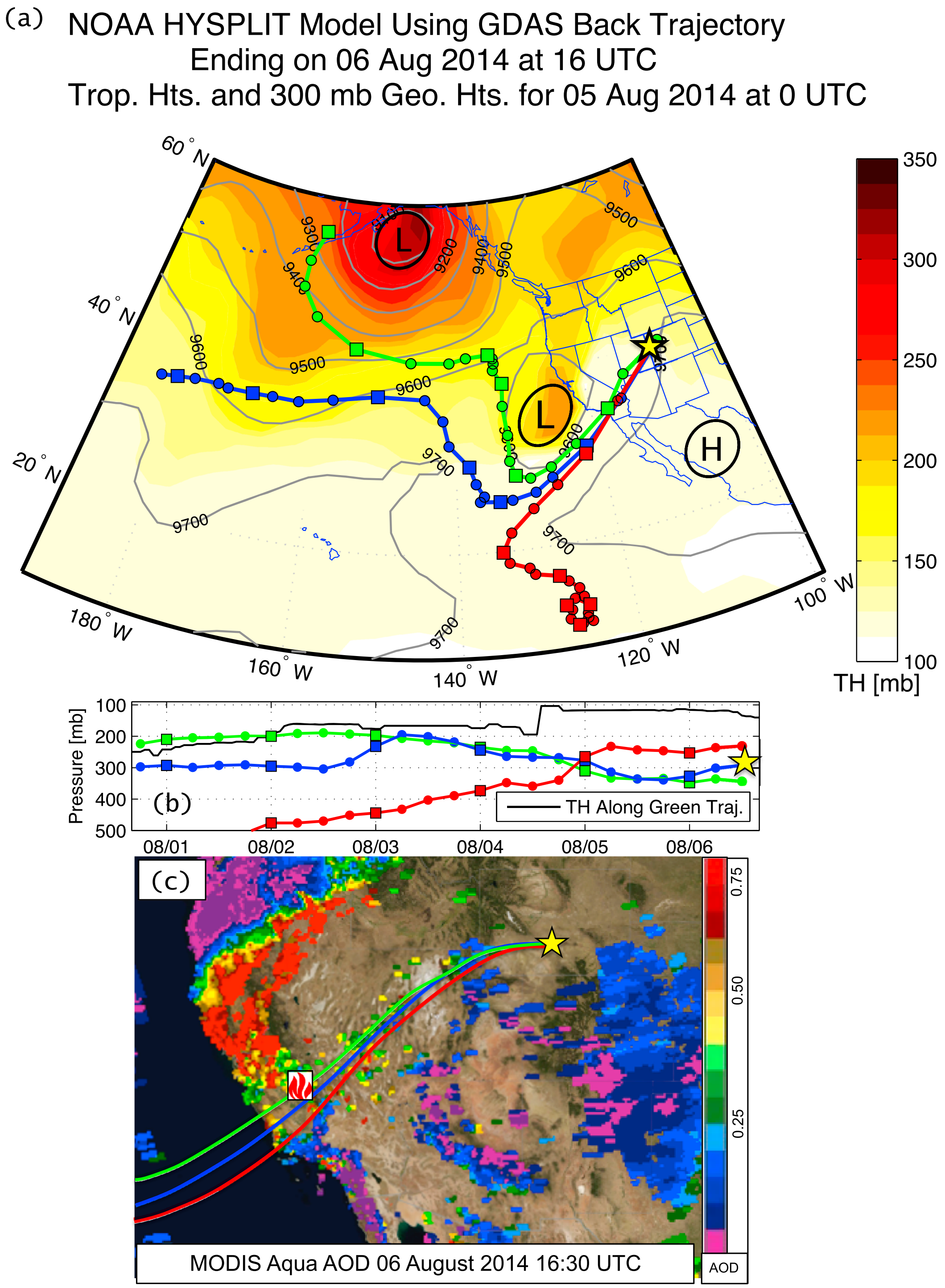

Figure 3. (a) Three 144 h back trajectories from the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model [*Draxler and Rolph*, 2003] using Global Data Assimilation System meteorological data starting at the Fort Collins, CO, site location, denoted with a black star. NCEP/NCAR Reanalysis geopotential height contours at 300 mb and the tropopause heights on 5 August 2014 at 0 UTC are shown. The trajectories are all initialized on 6 August 2014 at 16:00 UTC with starting pressure levels indicated by the red (230 mb/11.5 km asl), blue (300 mb/9.6 km asl), and green (360 mb/8.3 km asl) stars on the TROPOZ time series in Figure 1. (b) The vertical locations of each of the trajectories and the tropopause height (black line) above the green trajectory. (c) The trajectories overlaid on the Moderate Resolution Imaging Spectrometer (MODIS) Terra retrieved Aerosol Optical Depth (AOD) values for 6 August 2014 at 16:30 UTC (local 10:30 A.M.) (Source: NASA WorldView). A fire icon indicates the location of California's French Fire.

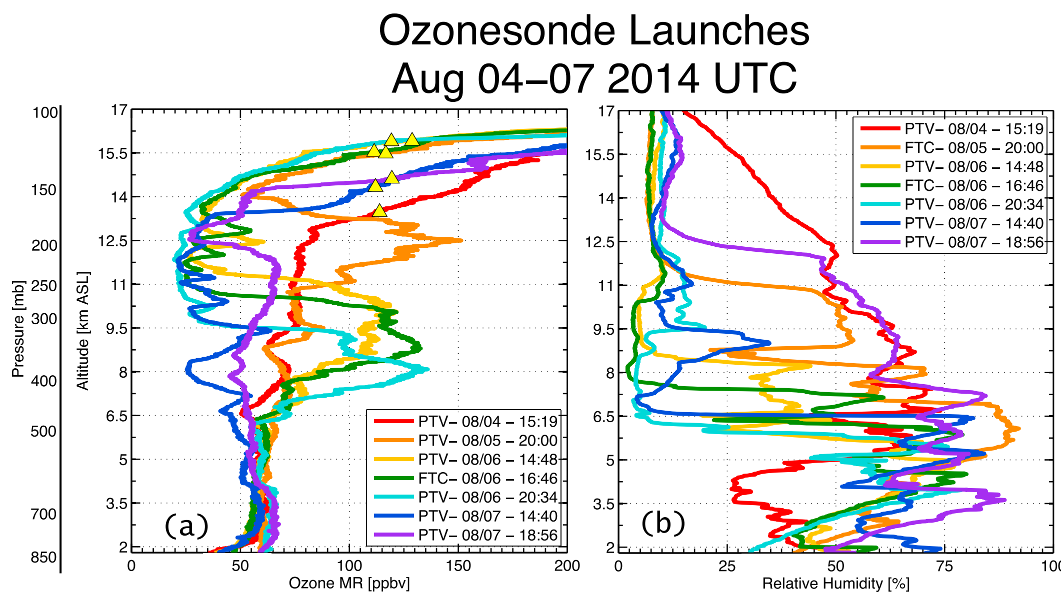

Figure 4. The (a) ozone and (b) relative humidity (RH) soundings illustrate the evolution of the STE event as it enters, subsides, and exits the Rocky Mountain region from 4 to 7 August 2014. Profiles from the Fort Collins-West (FTC) site and the Platteville (PTV) site (40.18°N, −104.73°W, 1523 m asl), roughly 40 km southeast of Fort Collins, are also shown. The ozone tropopause heights are indicated with yellow triangles.

[*Kalnay et al.*, 1996] tropopause heights for the green trajectory are represented with the black line in Figure 3b and indicate that the air parcel originated in the lower stratosphere, centrifugally detached from the outer edge or flank of the cyclone, and then resided below the tropopause height after 0 UTC on 2 August 2014.

Near 5 August 2014, all the trajectories show signs of deformation and cyclonic rotation around the cutoff low off the coast of California. This region of the Pacific Southwest coast has been previously identified as an area of high occurrence during a multidecadal climatology of cutoff low systems in the Northern Hemisphere that was constructed using 41 years (1958–1998) of NCEP/NCAR reanalysis data [*Nieto et al.*, 2005]. This study found that cutoff lows are much more common in the summer for this region and occur 49.5% of days sampled. All of the air parcels were shifted to the leading edge of the cyclonic system and each of the trajectories appears to maintain their respective altitudes. This meteorological system helped to stratify the air mass within troposphere during the southwesterly advection over the Pacific Southwest U.S. and into the Rocky Mountain region.

The largest Moderate Resolution Imaging Spectroradiometer (MODIS)-Terra aerosol optical depth (AOD) values in Figure 3c were observed in the Pacific Northwest, but there are also increased values directly near in the trajectory path near central California, Utah, and Nevada. The MODIS-Terra Fire Anomalies Product [*Justice et al.*, 2002] indicates a fire source location (37.275°N, −119.337°W, 1187.4 m asl) directly within the joint back trajectory region that is likely producing the increased AOD values. This wildfire source corresponds to California's French Fire, which burned more than 13,000 acres within the Sierra National Forest near Mammoth Pool. This fire, which began on 28 July 2014, was only 60% contained by 5–6 August 2014 (Source: California Department of Forestry and Fire Protection and the National Wildfire Coordinating Group (http://inciweb.nwcg.gov/incident/4013/)). During this time, the air mass was advected directly over the fire location, which suggests that this is a likely source for the aloft aerosol backscatter in the HSRL observations that resided below the TROPOZ observations of the ozone filament.

2.4. Balloon-Borne Soundings

Ozone (Figure 4a) and relative humidity (Figure 4b) profiles taken at the Fort Collins-West (FTC) and Platteville (PTV) sites during the DISCOVER AQ period provide context for the TROPOZ observations from 4 to 7 August 2014. The ozone soundings were made using Droplet Measurement Technology (DMT/EN-SCI, model 2Z-V7) instruments as described in *Martins et al.* [2013] and *Thompson et al.* [2014]. These sondes typically yield a precision better than ±(3–5)% and an accuracy of about ±(5–10)% [*Smit et al.*, 2007].

Ozonesonde profiles in Figure 4a are mostly between 50 and 60 ppbv below 6.5 km asl, and this discussion will focus on the STE event in the upper free troposphere. The red profile on 4 August 2014 at 15:19 UTC shows the vertical distribution of ozone before the STE event, in which most of the tropospheric ozone profile is centered around 60 ppbv and the ozone tropopause height (yellow triangle) is near 13 km. The orange profile on 5 August 2014 at 20:00 UTC, nearly 21 h before the TROPOZ observations began, shows the first signs of the stratospheric intrusion at the campaign sites, reaching 150 ppbv at 12.5 km. Although the potential temperature profiles are not shown, the anomaly in the stratospheric intrusion region (11 to 13 km asl) compared to the 4 August 2014 sounding is near −4 K and almost +6 K in the tropical low-ozone region (near 14 km asl).

The gold (PTV, 14:48), green (FTC, 16:46), and cyan (PTV, 20:34) profiles are all from 6 August 2014 and coincide with the TROPOZ observations. These soundings indicate downward motion of the high-concentration ozone reservoir, reaching nearly 125 ppbv between 7 and 11 km, depending on the launch time. These profiles also illustrate the tropical low-ozone region from 9.5 to 14 km and the persistently elevated tropopause height near 15.5 km. The dark blue and purple profiles indicate the free-tropospheric ozone concentrations mostly return to concentrations between 40 and 60 ppbv, but the tropopause height is still elevated with respect to the 4 August sounding.

Figure 4b shows the evolution of the intrusion event from the perspective of relative humidity, which appears to have a local minimum in the high-ozone regions and larger values in the low-ozone regions. The red profile on 4 August 2014 at 15:19 UTC shows the vertical distribution of relative humidity which varies mostly between 25% and 75% before the STE event reached the Rocky Mountain region. This sounding reveals increased midlevel moisture which was related to early morning cloud cover and a few isolated thunderstorms driven from upslope flow patterns.

The orange profile on 5 August 2014 at 20:00 UTC shows the initial intrusion of the stratospheric reservoir with dry air, denoted by RH mostly below 10% above 11 km. This 10% is indicative of the tropical air mass that has been transported within the upper troposphere for several days. The gold (PTV, 14:48), green (FTC, 16:46), and cyan (PTV, 20:34) profiles from 6 August 2014 illustrate the downward motion of the dry air intrusion associated with the stratospheric air. These profiles also begin to characterize the increase in relative humidity directly above stratospheric air mass associated with the tropical air. During this day, several of the soundings recorded instantaneous relative humidities less than 5% inside the stratospheric air mass. The purple profile, on 7 August 2014 at 18:56 UTC, reveals the return of the typical afternoon convective moisture pattern below 8 km, and the intrusion laminae has nearly dissipated.

2.5. Atmospheric Infrared Sounder Retrieval

The Atmospheric Infrared Sounder (AIRS) instrument on board the Aqua satellite has global coverage twice daily and provides many atmosphere quantities, including ozone (Figures 5a–5d) and relative humidity (Figures 5e–5h) [*Barnet et al.*, 2003]. Figure 5a corresponds to the closest AIRS overpass during the time of the STE event off the Pacific coast of California, where the Aleutian Low consistently had a large reservoir of ozone. An anomalous amount of ozone in the estimated intrusion region is apparent in the developing cut-off low-pressure system from Figure 3 between −120°W and −130°W and 30°N and 35°N. The region of low ozone on the equatorial side of the intrusion is most likely correlated with the transfer of the tropical air mass described earlier. Figure 5b on 6 August 2014 07:30 UTC shows the beginning of the southwesterly horizontal transport of ozone via the jet stream across the Pacific Southwest region with concentrations between 100 and 125 ppbv.

Figure 5c on 6 August 2014 19:30 UTC shows the continued transport of the ozone as it enters the Rocky Mountain region. Although the AIRS swath did not cover the Rocky Mountain region during this time, this panel is the closest overpass time and corresponds to near the center time of the TROPOZ observations (Figure 1). The AIRS retrieval indicates the advection of a 100 to 125 ppbv air mass throughout the Pacific Southwest region. However, the retrieval does not extend these increased concentrations to the Rocky Mountain region and, therefore may not be sampling the stratospheric impact to the troposphere. Figure 5d on 7 August 2014 07:30 UTC depicts the return to background values of ozone between 60 and 80 ppbv at 300 mb.

Figure 5e illustrates a dry region of air which corresponds to the intrusion location. Figures 5f–5h indicate the horizontal transport of the dry air mass over the Pacific Southwest region and into the Rocky Mountain region. The dry air mass, which had relative humidities mostly below 20%, compares well with the sonde observations

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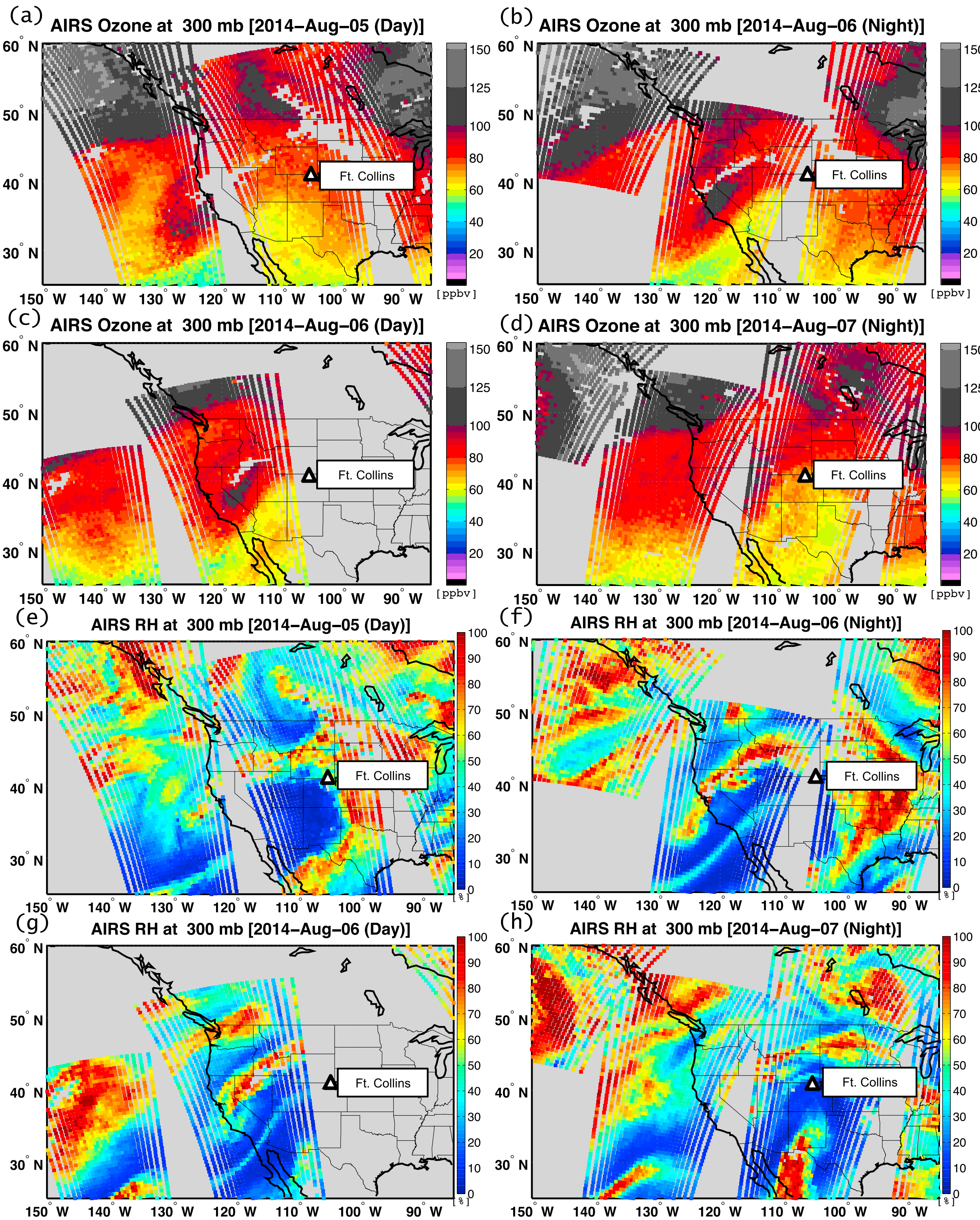

Figure 5. (a–d) Retrieved quantities of ozone at the 300 mb pressure level (the closest vertical level to the intrusion at Fort Collins). (e–h) The relative humidities. For the 5–6 August case study, the daytime retrieval (Day) occurs at 19:30 UTC and the nighttime retrieval (Night) occurs at 07:30 UTC. To ensure a quantitative analysis, only granules with 0 (high quality) and 1 (good quality) values are used in this work (Source : Goddard Earth Sciences Data and Information Services Center).

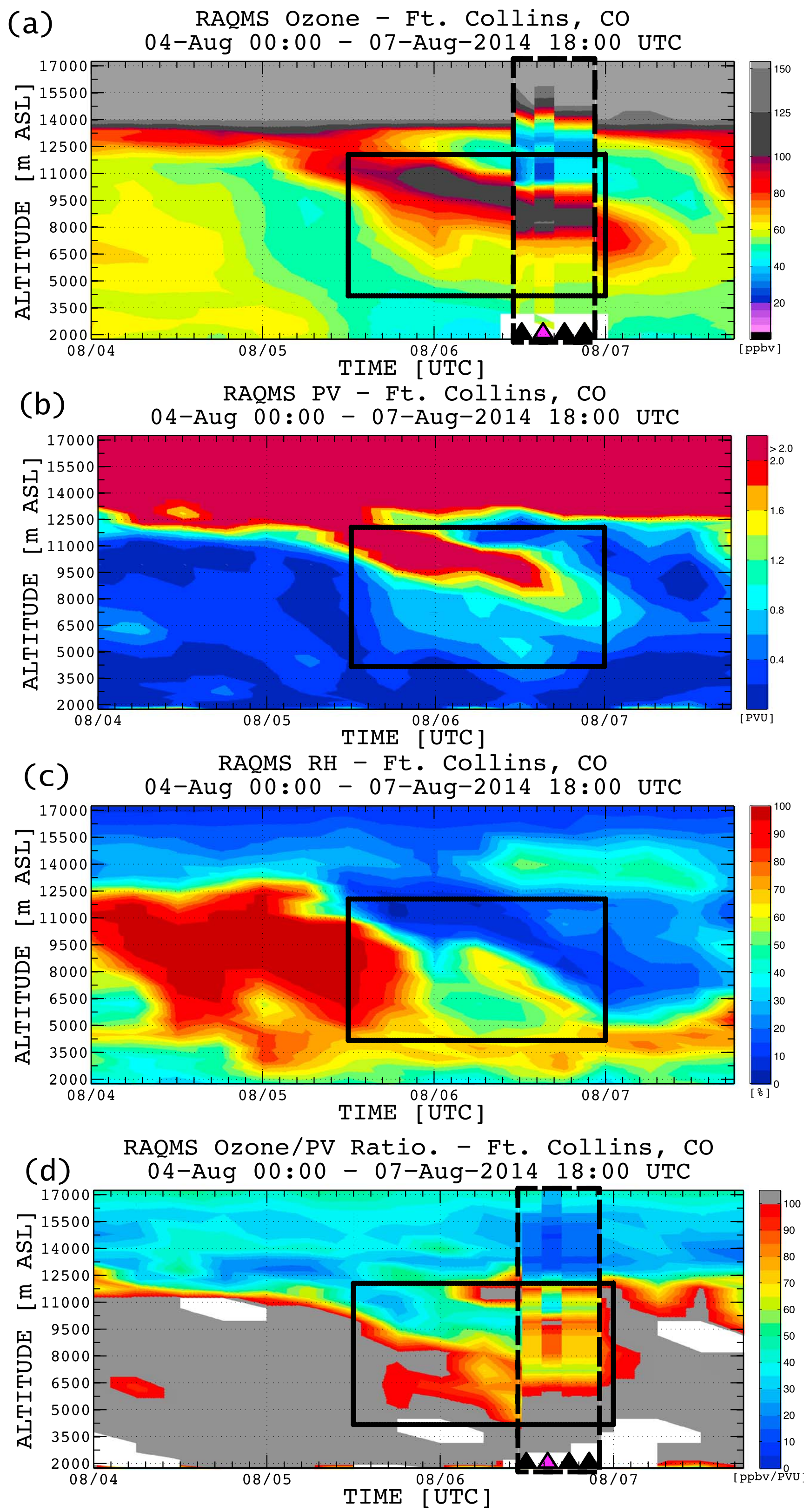

Figure 6. The Real-time Air Quality Modeling System (RAQMS) output shown as a time series at the Fort Collins location for (a) ozone, (b) potential vorticity, (c) relative humidity, and (d) the ratio of ozone to potential vorticity from 4 to 7 August 2014. The intrusion region is denoted with the black box, and the coinciding TROPOZ (black triangles) and ozonesonde (magenta triangle) observations are within the black dashed box.

from Figure 4. The thin line of tropical humid air below the cutoff low-pressure system near the intrusion site persists throughout the advection of the air mass.

2.6. RAQMS Model Outputs

To help investigate the August STE case study, the Real-time Air Quality Modeling System (RAQMS) output is shown as a time series from the closest model grid point to the Fort Collins site in Figures 6a–6d from 4 to 7 August 2014. RAQMS has simulated stratospheric air in the upper troposphere as compared to previous campaign observations [*Fairlie et al.*, 2007] and has been used to analyze deep stratospheric intrusions in the western U.S. [*Langford et al.*, 2015]. RAQMS is a unified (stratosphere/troposphere), online (meteorological, chemical, and aerosol) modeling system which has been developed for assimilating satellite observations of atmospheric chemical composition and providing real-time predictions of trace gas and aerosol distributions [*Pierce et al.*, 2007, 2009]. The RAQMS results here are based on a 1°×1° assimilation of cloud cleared total column ozone from Ozone Mapping Instrument and ozone profiles from Microwave Limb Sounder on the NASA Aura satellite. The daily assimilation cycle used here consists of 6 h online chemical/dynamical forecasts, initialized with NOAA Global Forecast System meteorological analyses at 00:00, 06:00, 12:00, and 18:00 UTC.

Output from RAQMS model at the Fort Collins site is shown for (a) ozone, (b) potential vorticity (PV), (c) relative humidity, and the (d) ozone to PV ratio. The solid black boxes correspond to the intrusion region, which were chosen to start in the center of the first full grid point in which PV values of 2.0 PV units (PVU, 1 PVU = 10^6 K m^2 kg^{-1} s^{-1}) occurred below the RAQMS dynamical tropopause height, which is described in the following section.

RAQMS ozone in Figure 6a reproduces the intrusion of an ozone-rich filament occurring on 5 August 2014 centered at 12:00 UTC, which corresponds well to 6 h after the estimated intrusion from the HYSPLIT back trajectory (Figure 3). Unfortunately, there were no TROPOZ observations on 4–5 August 2014, but the ozonesonde profiles from Figure 4 on 4 August 2014 15:19 UTC show similar ozone concentrations in the upper free troposphere. On 6 August 2014, TROPOZ measurements from 15:00 to 21:00 UTC (black triangles) and the ozonesonde (magenta triangle) launched near 18:00 from Fort Collins are overlaid on the RAQMS ozone time series within the dashed black box. The TROPOZ segments used for this comparison are a profile average from 6 August 2014 13:20 UTC to 18:00 UTC and from 18:00 UTC to 22:30 UTC, and the ozonesonde profile was used from 16:46 UTC. The RAQMS output agrees well with the TROPOZ ozone and sonde profile that indicate concentrations of 100 to 125 ppbv from 7 to 10 km. After the observations, RAQMS indicates further descent and dissipation of the layer, similar to the HSRL observations (Figure 1c). RAQMS also simulates the anomalously low ozone tropical air (near 45 ppbv) above the stratospheric intrusion.

Figure 6b shows RAQMS PV, a standard tracer for stratospheric air intruding into the troposphere [*Danielsen*, 1968]. The upper level PV anomaly is positive with respect to surrounding upper free tropospheric air, consistent with the transfer of a stratospheric air mass [*Lamarque and Hess*, 1994]. The background tropospheric air that is not affected by the intrusion has PV values mostly less than 0.8 PVU, and the stratospheric filament corresponds to values exceeding 2.0 PVU. Figure 6c depicts the evolution of the relative humidity during the intrusion event. The very dry region in Figure 6c correlates well with the ozone-rich air mass in Figure 6a and the increase in relative humidity from the tropical air mass.

Browell et al. [1987], *Ancellet et al.* [1994], *Beekmann et al.* [1994], *Stohl et al.* [2000], *Rao et al.* [2003], and *Kuang et al.* [2012] use the ratio of ozone to PV (Figure 6d) as a functional stratospheric tracer. *Stohl et al.* [2000] have shown that using PV only as a tracer can lead to large overestimations of STE events. We also employ the ozone to PV ratio as tracer for the Rocky Mountain region because it filters out ozone events that may be correlated with wildfire influences that are not associated with stratospheric dynamics [*Jaffe and Wigder*, 2012]. Constant values between 30 and 80 ppbv/PVU have been used to parameterize tropopause fold and intrusion events in various Northern Hemisphere locations [*Browell et al.*, 1987; *Beekmann et al.*, 1994]. However, as seen in Figure 6d, temporal information about the lifetimes of an STE event within the troposphere may be extracted from the variability of the ozone to PV ratio.

Near the initial stratospheric intrusion in Figure 6d, the ozone to PV ratios are mostly between 20 and 40 ppbv/PVU. As the stratospheric filament region dynamically evolves vertically and temporally, the ratios tend to increase toward ozone to PV ratios that are above 100 ppbv/PVU, which are consistent with typical tropospheric air values. This implies that in the region below the tropopause, an air mass with a low ozone to PV ratio correlates well with being near a recent influx of stratospheric air and a region of a large ozone to PV ratio value correlates well to photochemically produced ozone. Large ratios can also imply that the STE event has dissipated and mixed enough with the surrounding free troposphere, and it may be difficult to separate from background ozone. These trends will be used in the following section to help define a quantitative criteria for determining the amount and temporal evolution of stratospheric air below the tropopause.

3. Results

3.1. Identifying Stratospheric Air Within the Troposphere

From 4 to 08 August 2014, RAQMS simulated an ozone filament which was similar in altitude and concentration to the TROPOZ and ozonesonde observations. To further extend the analysis into other seasons and years, RAQMS model output parameters are used to investigate the stratospheric contribution below the tropopause at Fort Collins and the Rocky Mountain region for each season of 2012, 2013, and 2014. The seasonally dependent Northern Hemispheric PV criteria from *Kunz et al.* [2011] of 2.6 PVU for winter, 2.0 PVU for spring, 3.2 PVU for summer, and 3.2 PVU for fall are used to define the tropopause heights. In order to determine which simulated air masses were related to stratospheric events, it was necessary to remove model output that was directly related to background photochemically produced tropospheric ozone. In order to quantify the baseline free troposphere, the mean 2014 RAQMS PV and ozone to PV ratios were calculated near 300 mb as 0.78 PVU and 99.7 ppbv/PVU. For this reason, any RAQMS output with PV less than 0.8 PVU was removed to reduce contamination from boundary layer enhancements not pertaining to stratospheric air. Additionally, any RAQMS output below the tropopause that had higher-ozone to PV ratio values than 100 ppbv/PVU was denoted as background tropospheric air and removed. By following this ozone to PV ratio criteria, the most representative stratospheric contribution below each seasonally averaged tropopause height from 2012 to 2014 is estimated.

Figure 7a suggests that stratospheric air is responsible for 18–31% of tropospheric ozone throughout 2012–2014 at Fort Collins. The largest stratospheric contribution to the troposphere at Fort Collins tends to occur near the warmest dynamical tropopauses (Figure 7b); however, ozone reservoir abundances (Figure 7c) are still an important parameter. Because ozone is a greenhouse gas, larger amounts of ozone that reside near the tropopause, even well-mixed amounts, have resulted in elevated tropopause heights and may be important for decadal climate studies [*Santer et al.*, 2003].

Furthermore, these results indicate that the warmer temperatures allow for greater contribution of stratospheric air to be dynamically transferred into the troposphere. A similar seasonal behavior found in previous partial climatologies [*Price and Vaughan*, 1992; *Kentarchos and Davies*, 1998] was attributed to the existence of weaker jet streams during summer and fall, which allowed greater meridional flow and more likelihood in the probability of cutoff low formation. The life cycle of cutoff lows is dependent upon the temperature gradient from the cold air within the cutoff low and its surroundings. A much larger differential diabatic heating rate in the lower stratosphere during the summer/fall period than during the winter/spring period [*Dopplick*, 1972] also exists, which may generate an increase of PV values associated with STE events during the summer/fall period [*Ancellet et al.*, 1994]. Overall, this implies a stronger gradient (i.e., warmer upper air temperatures from increased stratospheric ozone to the upper troposphere) may prolong the duration of cutoff low-pressure systems and subsequently increase the duration and contribution of stratospheric air in the troposphere.

3.2. Determining the Lifetimes of Stratospheric Intrusions

The relative stratospheric contribution to Fort Collins, CO, for several seasons and years has been characterized in Figure 7a; however, it is also important to estimate the duration of the STE event below the tropopause. This has been performed with the summer 2014 RAQMS simulation to complement the DISCOVER AQ campaign observations presented in Figure 2. Because deeper intrusions often occupy the troposphere for a longer duration [*Stohl et al.*, 2003], it is important to quantify the seasonal contribution of STE events, as well as their lifetime within the troposphere. To estimate this, a linear regression is applied to mean ozone to PV ratio values within the August 2014 intrusion region (black box discussed with Figure 6d) and is shown in Figure 8a. This yields a dissipation rate of 0.91 ppbv/PVU/h, *y* intercept of 31.1 ppbv/PVU, $R^2 = 0.70$, and a root-mean-square error of 6.1 ppbv/PVU. The *y* intercept of 31.1 ppbv/PVU is in the initial stratospheric intrusion region ($t = 0$) and agrees well with previous work that indicates the annual mean ratio values are 30–45 ppbv/PVU in the lower stratosphere [*Beekmann et al.*, 1994; *Rao et al.*, 2003]. The triangles at 27 and 33 h are calculated with TROPOZ and ozonesonde ozone concentrations and corresponding RAQMS PV output. These triangles fall along the linear fit and are consistent with the RAQMS-only results. As the lifetime increases, the slope

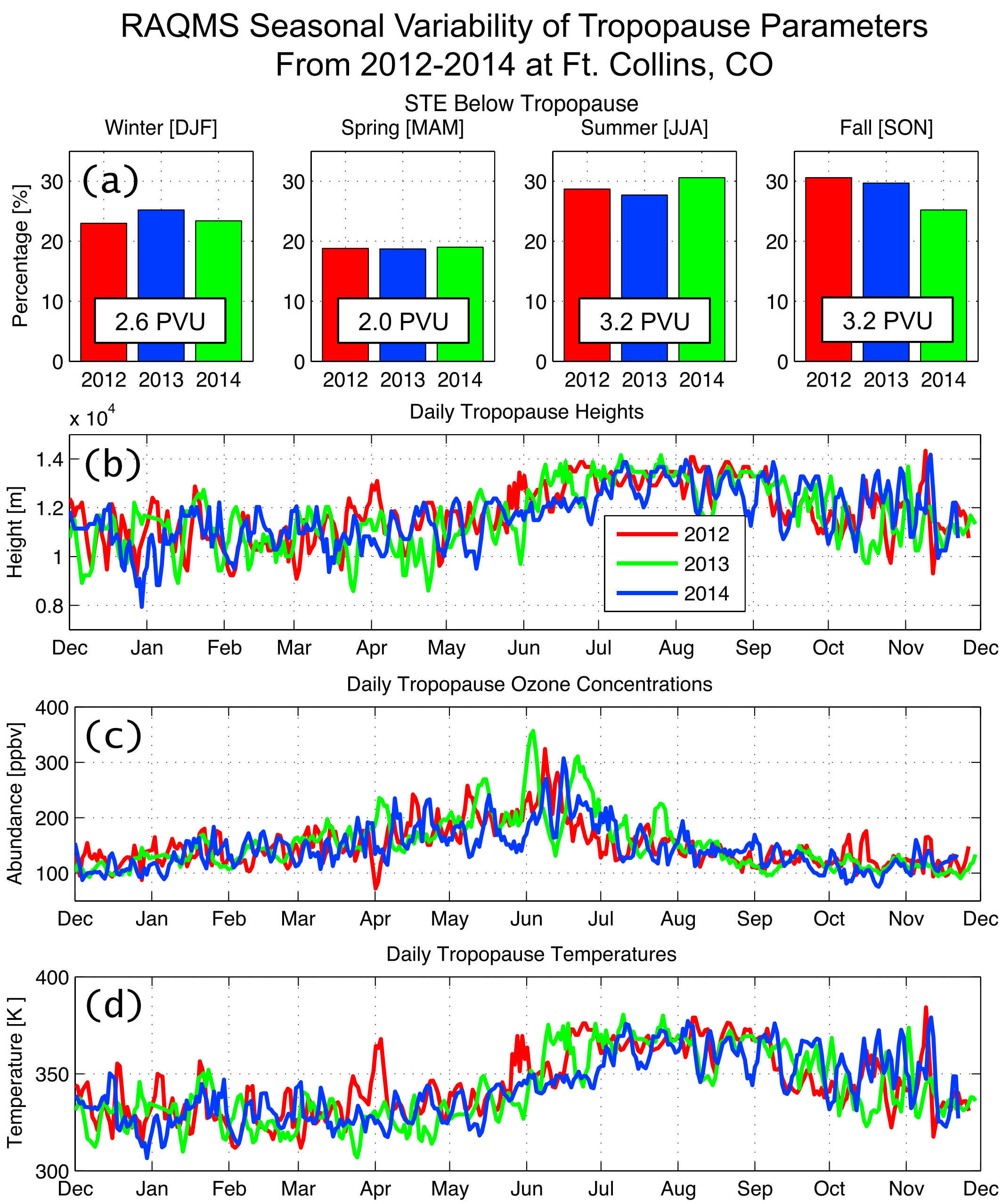

Figure 7. The seasonally dependent Northern Hemispheric PV criteria from *Kunz et al.* [2011] are used to define the (b) tropopause heights, which are an important parameter in determining the (a) amount of stratospheric contribution to the troposphere. The (c) ozone abundance and (d) potential temperature at the tropopause are also shown.

indicates that ozone to PV ratio values within the stratospheric intrusion increase toward values approaching those representative of background tropospheric ozone (over 100 ppbv/PVU).

Figure 8b extends the derived results for the STE lifetimes throughout the RAQMS output for summer 2014. To do this, the ozone to PV ratios are calculated at each RAQMS model point that met the previously described stratospheric air mass criteria for Figure 7a. Then, by using the dissipation rate derived for the summer 2014 STE event in Figure 8a, the duration of the stratospheric event below the tropopause is estimated. The most common lifetime of summertime stratospheric air below the tropopause is 12 h or less, which occurred 43% of the time. This suggests that almost half of the occurrences may have been quickly dissipating shallow stratospheric events or an extended lowering of the tropopause. Events with this duration are likely represented as the 10–30 ppbv increases in the free troposphere in first few kilometers below the tropopause height in

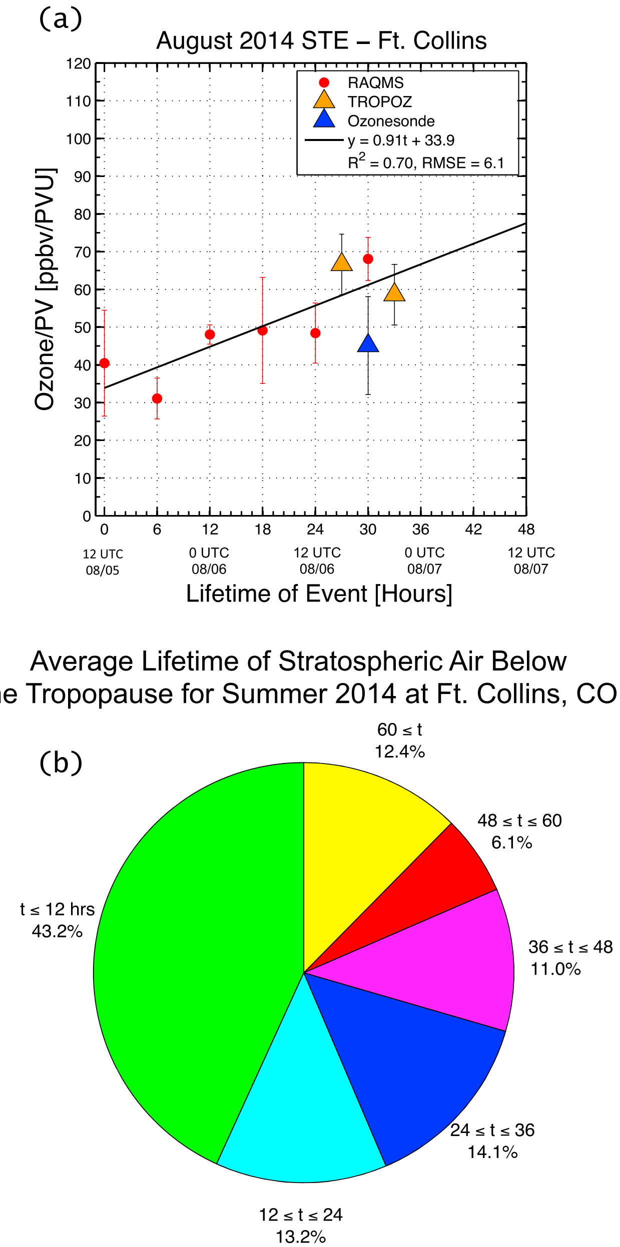

Figure 8. (a) The ozone to PV ratio and the correlation to its lifetime in the troposphere for the 4–7 August 2014 case. The RAQMS data in the figure represent the mean and standard deviation at each time step of the RAQMS data from Figure 6d after the intrusion. The TROPOZ and ozonesonde data also correspond to the mean and standard deviation of the measurements from Figure 6d. (b) The results from Figure 8a applied to RAQMS data for summer 2014.

Figure 2. It was also found that the sum of events lasting between 12 and 24 h (13.2%), 24 and 36 h (14.1%), and 36 to 48 h (11.0%) accounted for nearly 39% of all STE events. This implies that a significant fraction of the stratospheric air in the troposphere is correlated with multiday events that are likely to make it into the lower troposphere and be transported over large distances [*Langford*, 1999].

4. Conclusions

This paper has presented highly resolved observations and analyses of an intrusion of dehydrated, cold and ozone-rich stratospheric air into the upper free troposphere during NASA's DISCOVER AQ campaign in Colorado from 4 to 7 August 2014. The TROPOZ observations, in conjunction with the HSRL and sonde observations, have provided a rare look at the temporal evolution and downward motion of a stratospheric intrusion event. For the DISCOVER AQ campaign, TROPOZ observations indicate an increase in upper free tropospheric ozone concentrations, between 10 and 30 ppbv, which is largely contributed from recent stratospheric influx. For the case study presented, the stratospheric air, with ozone concentrations over 125 ppbv, intruded into the troposphere while being cyclonically rotated near an Aleutian Low and a cutoff low-pressure system near the Pacific coast of California. Back trajectories and AIRS retrievals illustrate the air mass being advected toward the Rocky Mountain region by an upper tropospheric jet stream.

To determine the overall impact of stratospheric air influx at Fort Collins, CO, during each season from 2012 to 2014, the RAQMS simulated ozone to potential vorticity ratio was used. Important seasonal parameters of RAQMS model output are presented, possibly for the first time in the Fort Collins, CO, region. The simulated seasonal variability indicates that the warming of the air near the tropopause is correlated with more frequent STE episodes. Results using the regression on simulated output for summer 2014 indicate that nearly half of the STE events ended within 12 h of the initial intrusion, indicating that shallow events are common in Fort Collins, CO, and this is consistent with the ozone increases from the extended campaign measurements. However, more than a third of the stratospheric air resided in the troposphere between 12 and 48 h, which signifies deeper intrusions into the lower free troposphere are also likely to occur. Although these conclusions are drawn from extended campaign measurements and model output, observations in other seasons, years, and regions will certainly strengthen and verify the calculated estimates of the occurrence and temporal variability of STE events.

Acknowledgments
Unless otherwise noted, all data used in this study can be found in the DISCOVER-AQ/FRAPPE data archive (http://www-air.larc.nasa.gov/missions/discover-aq), the TOLNet data archive (http://www-air.larc.nasa.gov/missions/TOLNet), or the RAQMS data archive (http://raqms.ssec.wisc.edu). This work was supported by UMBC/JCET (Task 374, Project 8306), the Maryland Department of the Environment (MDE, contract U00P4400079), NOAA-CREST CCNY Foundation (subcontract 49173B-02), and the National Aeronautics and Space Administration. The Platteville Nittany Atmospheric Trailer and Integrated Validation Experiment (NATIVE) operations were sponsored by NASA DISCOVER AQ grant NNX10ARG and the Pennsylvania State University. The University of Wisconsin HSRL operations were supported from the Colorado Department of Public Health and Environment (CDPHE). This research was supported by an appointment to the NASA Postdoctoral Program at the Goddard Space Flight Center. The authors gratefully acknowledge support provided by NASA HQ, the NASA Tropospheric Chemistry Program, and the Tropospheric Ozone Lidar Network (TOLNet). Thanks to the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and thanks to the Cooperative Institute for Mesoscale Meteorological Studies (CIMMS) for supporting the RAQMS model runs. Thanks to the helpfulness and expertise of Ryan Stauffer, Hannah Halliday, and Nikolai Balashov who worked with the NATIVE trailer at Platteville. Thanks to Debra Wicks Kollonige for providing her insight and recommendations on this work. Also, thanks to A.O. Langford for the extensive discussions on the heritage of stratospheric events. The views, opinions, and findings contained in this report are those of the author(s) and should not be construed as an official National Oceanic and Atmospheric Administration or U.S. Government position, policy, or decision.

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